

a minimum. The use of two insolubilizers means that only the minimum amount of reaction necessary to prevent caking of the granulated material prior to expansion will occur, thus leaving more silicate available for expansion, while still providing for a reaction at elevated temperatures in order to achieve water insolubility.

The resultant aggregate is quite lightweight, i.e., 1.5–25 pounds per cubic foot and, owing to the absence of organic materials, is completely fireproof. Likewise, depending mostly upon the nature of the insolubilizer used, the melting point of the aggregate is quite high, i.e., within the range of 1000°–2200° F. The aggregate, primarily non-load bearing in nature, is completely insoluble in boiling water and exhibits thermal conductivities on the order of 0.3 B.t.u. per hour per square inch of surface area per inch thickness per ° F. at a mean temperature of 75° F.

#### DESCRIPTION OF THE PREFERRED EMBODIMENTS

The term "aggregate" is used to describe the expanded material as a matter of convenience and for the reason that many of uses to which the material will be put correspond with the common uses of aggregate materials. The aggregate is in fact a porous, essentially spherical, particulate material of varying size depending upon such factors as the particle size of the ground material prior to expansion, the degree of expansion which the silicate material undergoes and the like. The aggregate may of course be provided in a shaped form. However, this is generally impractical once any significant cross-section is involved owing to the fact that the outer layer, which expands first, will serve to insulate the interior from heat, therefore preventing complete expansion and insolubilization.

The alkali metal silicates useful are sodium and potassium silicates having the stated weight ratios, preferably, and often hereinafter, sodium silicates having weight ratios within the range of 1:3.2–3.3, these being the materials most readily available commercially. The alkalinity of silicates having weight ratios of less than 1:3.0 is too high to warrant partial use, such alkalinity prohibitively increasing the amount of insolubilizer required to give a product having the desired properties. Silicates with weight ratios in excess of 1:7.0 are likewise not generally useful since the amounts of silicate solids which may be incorporated with these highly siliceous materials is too low.

The ranges of weight ratios of from 1:3.0–7.0 and of total silicate solids of from 80–40 percent are respective, that is, 80 percent is the maximum total alkali metal silicate solids possible with a 1:3.0 ratio material, whereas 40 percent solids is the minimum possible with a 1:7.0 ratio. Those skilled in the art will understand that the amount of solids required to achieve a like viscosity is less with a more siliceous silicate. Thus, the present invention contemplates that sodium silicates having 1:3.3–3.4 weight ratios will be used within the range of 50–80 percent total solids whereas a sodium silicate having a ratio of 1:7.0 is useful within the range of 40–55 percent. This relationship being established, the amounts of solids useful with the intermediate ranges will either be apparent to, or may readily be established by, those skilled in the art.

Generally, as a matter of convenience, the liquid and anhydrous alkali metal silicates will have the same weight ratios, although such is not required, any ratio within the stated range being satisfactory for either. Likewise, the relative amounts of liquid and anhydrous silicates are not critical, except that a combined silicate solids content of from 80–40 percent is required. For example, since a typical aqueous sodium silicate having a 1:3.22 ratio has a solids content of 39.3 percent, it is apparent that with 100 grams of such a solution there must be combined 57 grams of anhydrous silicate to obtain a 60 percent solids content.

The primary advantage of the use of this anhydrous silicate to increase the viscosity of the silicate-containing composition is that, while anhydrous silicate itself will not expand, admixture with the liquid silicate and the subse-

quent curing operation cause the anhydrous material to become hydrated and hence subject to subsequent expansion. Thus the purpose of providing a silicate composition which may be ground to a particulate form prior to expansion is served, while increasing, rather than decreasing as in certain prior art techniques, the amount of silicate capable of undergoing expansion.

The first step involves mixing the anhydrous with the liquid silicate, which is best done at temperatures in excess of ambient but less than boiling. Conveniently, the liquid silicate is first heated to a temperature on the order of 160° F. at which time admixing of the anhydrous silicate is commenced. Moisture loss becomes a consideration at temperatures substantially in excess of 160° F., although it may be avoided by the use of closed containers. At the boiling point premature foaming commences and is to be avoided.

The "primary insolubilizers" are those capable of reacting, for the most part, with the alkali portion of the silicate at temperatures less than boiling. Exemplary of these insolubilizers are boric acid; sodium, potassium, calcium, magnesium and zinc silicofluorides; alum; ferrosilicon, and calcium, magnesium and barium chlorides. The amount to be used varies with the identity of the insolubilizer and the weight ratio and quantity of the silicate used. A sufficient amount must be provided to prevent caking of the composition, caused by the hygroscopic nature of the alkali metal silicates, prior to expansion but subsequent to grinding. This amount may be lessened somewhat if the time lapse between grinding and expansion is short and excessive exposure to moisture at this stage is not contemplated. On the other hand, use of excessive quantities of the primary insolubilizer will reduce the silicate's capacity to expand. For example, in the case of  $\text{Na}_2\text{SiF}_6$ , with a sodium silicate having a 1:3.22 weight ratio and 60 percent silicate solids, from 5–15 percent by weight is effective.

The secondary insolubilizers useful in the practice of the present invention are those capable of reacting for the most part with the silicate portion of the alkali metal silicate at the temperatures involved in the expansion operation, i.e., 800–1800° F. The secondary insolubilizers are essentially insoluble in and nonreactive with the alkali metal silicate solution at temperatures less than 250° F. Exemplary of the secondary insolubilizers are magnesium phosphate, tribasic; portland cement; calcium silicate; iron and zinc oxides; calcium, barium and magnesium carbonates, and fly ash. The amount used is that amount sufficient to render the expanded aggregate water-insoluble, it having been noted that less than the amount theoretically required for complete reaction with silicate will achieve this effect. For example, using calcium carbonate as the secondary insolubilizer it has been found that an aggregate having, for all practical purposes, complete water insolubility may be produced by the use of only 75 grams of calcium carbonate for every 100 grams of sodium silicate solids (silicate weight ratio 1:3.22), 127 grams of calcium carbonate being the stoichiometric quantity. The expense of the insolubilizer generally being greater than that of the silicate and the presence of large quantities tending to increase the density of the resultant aggregate, it becomes apparent that the use of minimum amounts of these materials is usually desirable.

The total amount of insolubilizers, both primary and secondary, is generally considerably less than the stoichiometric amount required for total reaction with the alkali metal silicate present. For example, using a sodium silicofluoride-calcium carbonate insolubilizer system, it has been found that the use of from 21–84 percent of the stoichiometric amount results in an aggregate having the desired properties. Use of larger quantities acts primarily to increase the cost of the aggregate as well as to increase its density (by reducing the degree of expansion obtainable and by acting as a filler). On the other extreme, too little of these materials results in caking of the ground material and/or a water-soluble aggregate,